OPTICAL AND PROBE DIAGNOSTICS OF PLASMA-LIQUID SYSTEMS WITH SECONDARY DISCHARGE

I.V. Prysiazhnevych, V.Ya. Chernyak, E.K. Safonov

Department of Physical Electronics, Radiophysical Faculty,
Taras Shevchenko Kyiv National University, Pr. Acad. Glushkova 2/5, 03127 Kyiv, Ukraine,
e-mail: priv@ukr.net, chern@univ.kiev.ua

A low-pressure plasma-liquid system with secondary discharge was investigated by optical and probes techniques. Emission spectra at the different cross-sections of plasma-liquid reactor were analyzed and the electronic excitation temperature was determined. The voltage jump near the plasma-liquid boundary was observed and possibility of varying of its value by changing the voltage on the secondary discharge was shown.
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1. INTRODUCTION

Plasma is known to be chemical active medium with high concentrations of reactive components. However, the price for such high activity of plasma is multi-channel passing of chemical reactions and weak control of this process. It was generally supposed that the main way to increase a selectivity of plasma-chemical transformations is transferring from thermal to non-equilibrium plasma. However, gas-discharge sources of non-equilibrium plasma can guarantee only plasma non-isothermality (when the temperature of electrons differs from characteristic temperatures of heavy particles) under average electron's energy lower than energies corresponding to the maximum of the cross-sections of excitation for inelastic processes. Furthermore, the electron temperature varies in the narrow range even under considerable changes of external parameters of the gas discharge (current, voltage) due to the exponential dependence of the ionization rate on the electron's temperature. The last remark is pertinent to the volume of plasma, but not to the region of plasma-liquid or plasma-solid contact. That is why the two-phase gas-liquid plasma systems with one or two electrodes immersed into the liquid are of great interest today [1-3].

The main peculiarity of plasma-liquid systems is an intensive molecule's flow emitted from the free liquid surface into the discharge volume due to both the solution evaporation and the formation of gaseous plasma-chemical products. Low-pressure conditions for plasma-liquid system provide an effective contact of plasma with liquid surface. To maintain pressure inside system almost constant the volume of plasma-chemical reactor was continuous pumped. However discharge burning under pressures ~10 Torr (corresponds to the right (ascending) branch of the Paschen's curve) and presence of negative ions in plasma-liquid systems with gaseous discharge promote development of overheating plasma instability both [4]. It is clear that development of instabilities in such system in no case can promote solving the selectivity problem. Using of auxiliary discharge inhibits development of the ionization instability in plasma of the secondary discharge, provides high uniformity of parameters on the plasma-liquid boundary and increases a number of external parameters of influencing on the plasma-liquid interaction in real plasmachemical process. Therefore, the secondary discharge with one "liquid" electrode supported by means of plasma flow of the auxiliary discharge is of great interest today [5-7].

2. EXPERIMENT

The scheme of low-pressure plasma-liquid system with secondary discharge is shown on Fig.1.

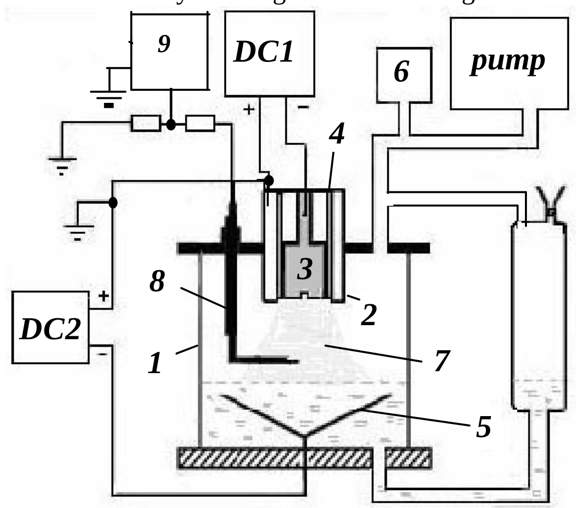

Fig.1. Schema of low-pressure plasma-liquid system.

It consists of the cylindrical quartz chamber *1* (of 80-mm diameter and 160 mm of height) partially filled by liquid; source of the auxiliary discharge (coaxial-end discharger with stainless steal anode *2*, hollow cathode *3* and glass tube *4,* which separates anode and cathode); funnel-shaped stainless steal electrode *5* (with diameter of cone base ~ 6 cm). Such construction of the electrode decreases instability of liquid surface caused by gaseous products of electrolysis occurring on this electrode. Exhaustion of the chamber was made by mechanical vacuum pump, pressure inside system was measured by vacuum meter *6*.

The voltage for the secondary discharge U_d was applied between the metallic electrode *5* immersed into a liquid and anode *2* of the auxiliary discharge (*DC2* on Fig.1). The auxiliary discharge was powered by *DC1* source (Is, Us– current and voltage on the auxiliary discharge correspondingly).

Distilled water and NaOH solutions (with different concentrations 0,001-0,1 mol/l) were used as working liquids. The distance between anode of the coaxial-end discharger and liquid surface did not exceed 20 mm, while the height of the liquid column above the electrode *5* was ~ 15 mm. The probe technique was used to measure the axial distributions of the floating potential in the plasma-liquid system. The molybdenic probe *8* (diameter ~ 300μm, length ~ 1mm) was connected via the divider ($4\times10^7\Omega$/ $2\times10^4\Omega$) directly to the oscilloscope *9* (Fig.1). The probe was moving along to the axis of the chamber.

Method of optical emission spectroscopy (OES) was applied for optical diagnostics. The emission spectra of plasma were observed in the range of wavelength 220-1100 nm (with spectral resolution 0.3 nm) by CCD-based multi-channel UV-VIS-NIR optical spectra analyzer (MOSA).

3. RESULTS AND DISCUSSION

After degassing of the liquid, the auxiliary and the secondary discharges were switched on. The both modes (different polarities of the immersed into the liquid electrode: negative -"liquid" cathode, and positive - "liquid" anode) were investigated. It was observed that plasma column *7* of the secondary discharge (Fig.1) has a truncated cone-shape with extension at the liquid surface.

Current-voltage characteristic (VAC) of the secondary discharge in plasma-liquid system was measured. The VAC is given on Fig.2.

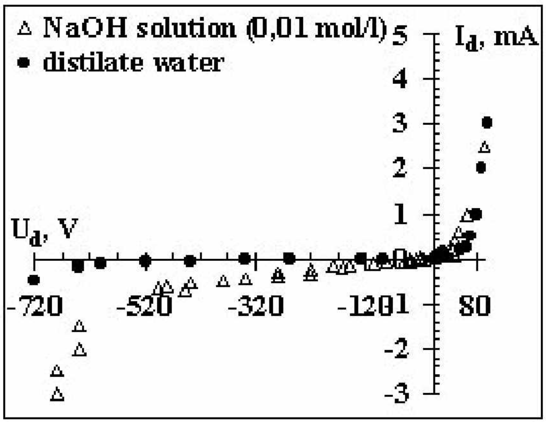

Fig.2. VAC of the secondary discharge in plasma-liquid system (pressure is ~15 Torr, I_s=200 mA, U_s=450 V)

From Fig.2 can be seen, that VAC of the secondary discharge in plasma-liquid system consists of several typical zones: positive exponential branch and negative branch, which contains the line section and the range corresponding to the abrupt exponential current increasing. It was shown that its behavior is similar to usual current-voltage curves of the Langmuir probe in the range of low currents, while in the range of large currents investigated VAC has a sharp growth character, as for the secondary gas discharge in strong electric fields [4].

The axial distributions of the potential on the secondary discharge in plasma-liquid system with a liquid electrode are represented on Fig.3.

The voltage jump near liquid surface was observed. Such voltage increasing at the plasma-liquid contact can be interpreted as result of conductivity decreasing of plasma near the liquid surface due to the high concentration of negative ions at that region [6].

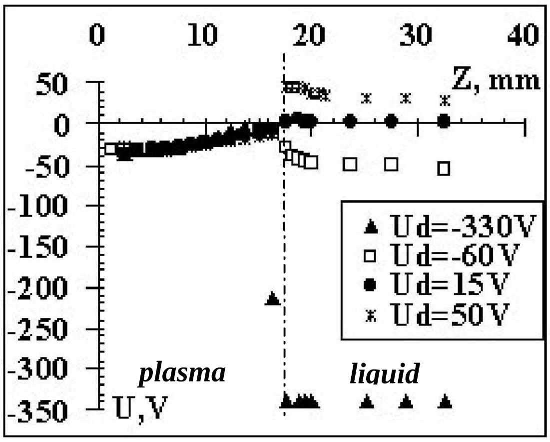

Fig.3. Axial distribution of floating potential in plasma-liquid system with NaOH solution (concentration 0,01 mol·l^{-1}, pressure ~ 10 Torr, I_s=200 mA, U_s=450 V).

At the same time, the potential jump near plasma-liquid boundary can be associated with the basic peculiarity of the potential distribution surround electrode immersed into the plasma. The behaviour of VAC of the secondary discharge at the range of low currents, which is similar to the VAC of the Langmuir probe, justifies this conclusion. From Fig.3 seen, that it is possible to vary the voltage jump value by changing the secondary discharge voltage. It was shown that its amplitude is proportional to the voltage on the secondary discharge (Fig.4).

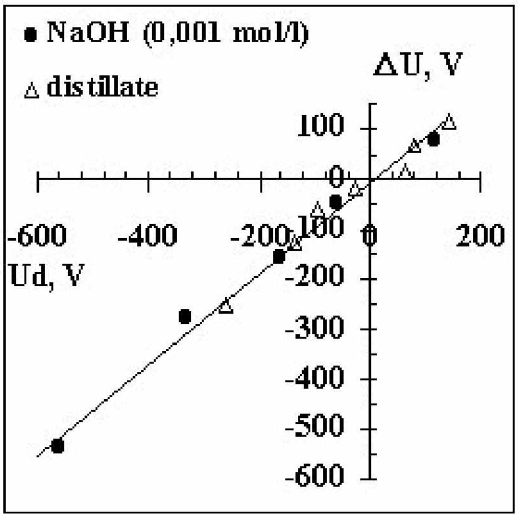

Fig.4. Dependence of the potential jump ΔU on the voltage on the secondary discharge U_d.

(Pressure ~15 Torr, Is=200 mA, Us=450 V).

Thus, the analogues between low-pressure plasma-liquid system with secondary discharge system and single probe were revealed. The existence of voltage jump near liquid surface (as near a single probe) lets to consider the liquid surface as electrode, immersed into the plasma, and to claim that the main reason of its appearance is shielding of the liquid surface by the spatial charge of the corresponding polarity.

The optical diagnostics of low-pressure plasma-liquid system with the secondary discharge has shown that the most intensive spectral lines and bands in emission spectra (obtained after degassing of the system) belong to hydrogen H (H_α 656.3 nm, H_β 486.1 nm, H_γ 434.0 nm), oxygen O (777.3, 844.6, and 926.0 nm) and hydroxyl OH (0,0-band at 306-309 nm). Fast response of used MOSA (min exposure time ~7 ms) allowed to explore temporary changes of emission H lines and OH bands intensities during the discharge burning within the time t= 1-30 min. It was found that the intensity of H_α line is higher near liquid surface than at the 5 mm distance from anode *2* (Fig.1). It is almost constant during plasma treatment in the case of "liquid" anode and rapidly decreases with increasing t in the case of "liquid" cathode at the section near the source of the auxiliary discharge. The electronic excitation temperature T_e^* was determined from the relative intensities I_λ of H_α, H_β lines. It was found that T_e^* values and its distributions in time differ for different sections of the plasma-liquid system. After t=15 min of plasma treatment T_e^*~0,8 eV (at the section 3-5 mm above the liquid surface) and T_e^*~0,5 eV (5 mm below anode *2* Fig.1) correspondingly [7].

4. CONCLUSIONS

- The VAC of the secondary discharge with a liquid electrode (namely its exponential positive and linear area of the negative branches) is similar to VAC of Langmuir probe.
- The nature of the potential jump near plasma-liquid contact deals with spatial charge shielding of the electrode of the secondary discharge immersed into the plasma flow of the auxiliary discharge. The possibility of varying its value by changing of the voltage on the secondary discharge was shown.
- The electronic excitation temperature T_e^* of H atoms was determined. The behavior of the H_α line intensity over the time of plasma treatment was shown to be different near the liquid surface and near the source of the auxiliary discharge.

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ОПТИЧЕСКИЕ И ЗОНДОВЫЕ ИССЛЕДОВАНИЯ ПЛАЗМОЖИДКОСТНОЙ СИСТЕМЫ НИЗКОГО ДАВЛЕНИЯ С ВТОРИЧНЫМ РАЗРЯДОМ

И.В. Присяжневич, В.Я. Черняк, Е.К. Сафонов

Оптической и зондовой методиками исследована плазможидкостная система низкого давления с вторичным разрядом. Проанализированы эмиссионные спектры плазмы в различных сечениях плазможидкостного реактора и определена температура заселения электронных возбужденных уровней. Наблюдается скачок потенциала вблизи границы плазма-жидкость, показана возможность управления его величиной за счет изменения напряжения на вторичном разряде.

ОПТИЧНІ ТА ЗОНДОВІ ДОСЛІДЖЕННЯ ПЛАЗМОВО-РІДИННОЇ СИСТЕМИ НИЗЬКОГО ТИСКУ З ВТОРИННИМ РОЗРЯДОМ

І.В. Присяжневич, В.Я. Черняк, Є.К. Сафонов

Оптичною та зондовою методикою досліджено плазмово-рідинну систему низького тиску з вторинним розрядом. Проаналізовано емісійні спектри плазми у різних перетинах системи плазмово-рідинного реактору та визначено температуру заселення електронних збуджених рівнів. Спостерігається стрибок потенціалу біля границі плазма-рідина, показано можливість керування величиною цього стрибка за рахунок зміни напруги на вторинному розряді.